

Reprinted from

ATMOSPHERIC ENVIRONMENT

Vol. 31, No. 4, pp. 529-544

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AREAS—INDOOR/OUTDOOR RELATIONSHIPS

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PERGAMON

1996

RWLP0251

PM3006519115



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(First received 3 February 1996 and in final form 5 July 1996)

Abstract—Air quality monitoring was carried out to collect data on the levels of various indoor and ambient air constituents in two cities in Korea (Seoul and Taegu). Sampling was conducted simultaneously indoors and outdoors at six residences, six offices and six restaurants in each city during summer 1994 and winter 1994–1995. Measured pollutants were respirable suspended particulate matter (RSP), carbon monoxide (CO), carbon dioxide (CO₂), nitrogen dioxide (NO₂), and a range of volatile organic compounds (VOCs). In addition, in order to evaluate the effect of smoking on indoor air quality, analyses of parameters associated with environmental tobacco smoke (ETS) were undertaken, which are nicotine, ultraviolet (UVP), fluorescence (FPM) and solanesol particulate matter (SolPM). The results of this study have confirmed the importance of ambient air in determining the quality of air indoors in two major Korean cities. The majority of VOCs measured in both indoor and outdoor environments were derived from outdoor sources, probably motor vehicles. Benzene and other VOC concentrations were much higher during the winter months than the summer months and were not significantly greater in the smoking sites examined. Heating and cooking practices, coupled with generally inadequate ventilation, also were shown to influence indoor air quality. In smoking sites, ETS appears to be a minor contributor to VOC levels as no statistically significant relationships were identified with ETS components and VOCs, whereas very strong correlations were found between indoor and outdoor levels of vehicle-related pollutants. The average contribution of ETS to total RSP concentrations was estimated to range from 10 to 20%. Copyright © 1996 Elsevier Science Ltd

Key word index: Volatile organic compounds (VOCs), respirable suspended particulate matter (RSP), benzene, environmental tobacco smoke (ETS), indoor air quality (IAQ), nicotine.

INTRODUCTION

Over the past two decades there has been a rapid increase in urbanisation and industrialisation in Korea. With this has come a dramatic increase in the number of residences, office buildings and manufacturing facilities, together with increases in both the number and density of motor vehicles. The industrialisation/urbanisation process has had both positive and negative effects on indoor air quality in Korea (Kim, 1992). Many of the newer office buildings found in Korean cities have several storeys and are equipped

with centralised air conditioning systems, including state-of-the-art air filtration devices. When operated properly, such equipment can produce substantial improvements in indoor air quality. But modern office buildings, including those in Korea, are not always operated and maintained properly. This has led to the emergence in Korea of "sick" buildings characterized by air quality problems that can be even more severe than those presented by ambient air (Lee *et al.*, 1995).

Industrial emissions and motor vehicles are believed to be the major causes of ambient air pollution in Korean cities (Baek, 1991). Emissions from these sources have immediate implications for the indoor environment, since the air in both naturally and mechanically ventilated buildings is replenished to

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varying degrees with ambient air, which may or may not be filtered or otherwise conditioned before being brought indoors. Several studies have demonstrated that ambient air can have a significant impact on the indoor environment (Yocom, 1982; Daisey *et al.*, 1994; Perry and Gee, 1994).

Still other pollution originates indoors, being traceable to factors such as building materials, office equipment, cleaning supplies and the occupants themselves. It is significant that many homes in Korea continue to rely upon coal briquettes as heating and cooking fuel, leading to elevated indoor carbon monoxide levels, particularly during cold winter months (Son *et al.*, 1990). Although coal briquettes are slowly being replaced by bottled propane and natural gas for cooking and heating, these replacements present their own indoor pollution problems when not handled in an appropriate manner. In addition, many Korean homes, offices, shops and restaurants use portable kerosene devices to provide supplementary heating during the winter months (Leslie and Haraprasad, 1993). When not vented properly, such devices can increase indoor pollution to extreme levels (Kodama *et al.*, 1991; Kawamoto *et al.*, 1993).

Although questions have been raised in Korea about air quality in underground shopping arcades, train stations and other public places, the air quality monitoring data that are available for those sites are not adequate to permit policy-makers to develop an accurate profile of the actual situation or to identify the major contributors to air quality problems (Korea Environmental Science Council, 1989; Cha and Cho, 1993). Moreover, very few studies have focused on other indoor environments such as homes, offices and restaurants, which can be expected to be the most important exposure sites for a substantial percentage of the Korean population.

Air quality monitoring was carried out in this study in two large Korean cities (Seoul and Taegu). Sampling was conducted simultaneously both indoors and outdoors at six residences, six offices and six restaurants in each city. Full sampling runs were completed at the 36 target sites on two separate occasions (summer 1994 and winter 1994/1995). Among other things, this approach permitted an assessment to be made of the contribution made by the ambient air to the quality of the air indoors, an insight that has been lacking in some of the other indoor air quality studies that have been undertaken in Korea.

Several air quality parameters were measured simultaneously in order to develop a general profile of both indoor and ambient air and to assess the relationship between them. The measured pollutants were respirable suspended particulate matter (RSP), carbon monoxide (CO), carbon dioxide (CO₂), nitrogen dioxide (NO₂), nicotine, and a range of volatile organic compounds (VOCs). In addition, in order to estimate the contribution of environmental tobacco smoke (ETS) to total RSP, three components related to ETS particles were analysed; ultraviolet-absorbing

particulate matter (UVP), fluorescing particulate matter (FPM) and solanesol-related particulate matter (SolPM). Although each of these methods has been evaluated individually, few intercomparisons have been made using field samples (Ogden *et al.*, 1990; Phillips *et al.*, 1994).

One way in which to classify the pollutants measured in this study is in their degree of specificity to a given pollution source. RSP and VOCs can emanate from a variety of sources, both combustion-related and otherwise. CO and CO₂ are indicative of combustion processes and/or human metabolism. FPM and UVP are indicators of the fraction of particulate matter derived from combustion and can be taken as an estimate of the upper limit of the ETS contribution to RSP levels. Nicotine (vapour phase) and solanesol (particle phase) are markers that are essentially specific to the burning of tobacco.

The main objectives of this study were (i) to provide quantitative information on the levels of potentially important pollutants in three typical Korean micro-environments (homes, offices and restaurants), (ii) to compare indoor and ambient pollution levels as part of the task of source apportionment, and (iii) to investigate the extent to which certain indoor pollution sources influence the quality of indoor air in urban areas in Korea.

MATERIALS AND METHODS

Sampling protocol

Information concerning the sampling sites selected in Seoul and Taegu are shown in Table 1. The population of Seoul, the capital of Korea, is more than 11 million. Taegu, the third largest Korean city with a population of 2.5 million, is located 300 km to the southeast of Seoul. Sampling was conducted during both summer (August to September 1994) and winter months (December 1994 to January 1995). Four sets of identical sampling equipment were assembled for the study, two in each city, operated by two teams of operators, who were always present throughout the sampling periods.

Prior to the main study, a number of pilot experiments were completed to confirm the suitability of the sampling and analytical procedures that had been proposed for the main study. These included the estimation of lower detection limits using actual field and blank samples, determination of appropriate sampling volumes, performance cross-checking between the four sets of samplers and an interlaboratory comparison for nicotine determination, involving Hanyang University and the Environmental Toxicology Laboratory of the Korea Research Institute of Chemical Technology.

At indoor locations, the sampling equipment was placed in the centre of the room at a height of approximately 1.5 m in order for the sampling to occur in the breathing zone of a seated person. Outdoor sampling locations were chosen to avoid significant point sources of pollution, such as building exhaust vents.

Samples were taken at 12 residences, 12 offices and 12 restaurants (six of each type in both cities). Each of the residences was occupied by a single family, and the residences were dispersed geographically throughout each city. An effort was made to balance the residences with respect to building type and heating and cooking practices. None had an attached garage. Roughly half of the residences were occupied by at least one active smoker, the others being

Table 1. Identification of sampling sites in Seoul and Taegu

Site	No.	Type	Floor	Size (m ²)	Cooking fuel	Heating type (fuel) ^a	Ventilation	City and area ^b	Kerosene stove use in winter	Air conditioning in summer	Smoking allowed
Home	1	Semi-western	Ground	99	LPG	Central (K)	Natural	Seoul (R)	Yes	No	Yes
	2	Semi-western	1st	83	LPG	Central (K)	Natural	Seoul (R)	No	No	No
	3	Semi-western	Ground	33	LPG	Central (B)	Natural	Seoul (R)	Yes	No	No
	4	Korean	Ground	43	LPG	Central (B)	Natural	Seoul (R)	No	No	Yes
	5	Korean	Ground	43	LPG	Central (B)	Natural	Seoul (C)	No	No	Yes
	6	Semi-western	1st	106	LPG	Central (L)	Natural	Seoul (R)	No	No	Yes
	7	Korean	Ground	100	LPG	Central (K)	Natural	Taegu (R)	No	No	No
	8	Korean	Ground	150	LPG	Central (B)	Natural	Taegu (R)	No	No	Yes
	9	Semi-western	Ground	130	LPG	Central (K)	Natural	Taegu (R)	Yes	No	No
	10	Semi-western	Ground	100	LPG	Central (L)	Natural	Taegu (R)	No	No	No
	11	Apartment	1st	90	LPG	Central (K)	Natural	Taegu (R)	No	No	Yes
	12	Apartment	2nd	109	Town gas	Central (L)	Natural	Taegu (R)	No	No	Yes
Office	1	Public	Ground	46	Not used	Central (K)	Mixed	Seoul (C)	No	Yes	Yes
	2	Private	2nd	40	Not used	Local (K)	Natural	Seoul (C)	Yes	Yes	No
	3	Private	1st	61	Not used	Local (K)	Natural	Seoul (C)	Yes	Yes	Yes
	4	Private	4th	61	Not used	Central (L)	Mixed	Seoul (C)	No	Yes	Yes
	5	Private	1st	30	Not used	Central (L)	Mixed	Seoul (C)	No	Yes	Yes
	6	Private	2nd	50	Not used	Central (L)	Mixed	Seoul (C)	Yes	Yes	Yes
	7	Bank	Ground	380	Not used	Central (L)	Mixed	Taegu (C)	No	Yes	No
	8	High School	1st	140	Not used	Local (K)	Mixed	Taegu (R)	Yes	No	No
	9	Public	Ground	190	Not used	Central (L)	Mixed	Taegu (C)	No	Yes	No
	10	Public	Ground	490	Not used	Central (L)	Mixed	Taegu (R)	No	Yes	Yes
	11	Private	3rd	270	Not used	Local (K)	Mixed	Taegu (R)	Yes	No	Yes
	12	Public	2nd	250	Not used	Central (L)	Mixed	Taegu (R)	No	Yes	Yes
Restaurant	1	Korean	Ground	165	LPG, charcoal	Local (K)	Mixed	Seoul (C)	Yes	Yes	Yes
	2	Korean	1st	215	LPG, charcoal	Local (K)	Mixed	Seoul (C)	Yes	Yes	Yes
	3	Korean	Ground	99	LPG, charcoal	Local (K)	Mixed	Seoul (R)	Yes	Yes	Yes
	4	Korean	Ground	83	LPG	Local (K)	Natural	Seoul (C)	Yes	Yes	Yes
	5	Korean	Ground	99	LPG, charcoal	Local (K)	Mixed	Seoul (C)	Yes	Yes	Yes
	6	Korean	Ground	30	LPG	Local (K)	Natural	Seoul (C)	Yes	Yes	Yes
	7	Korean	Ground	90	LPG, charcoal	Local (K)	Mixed	Taegu (C)	Yes	Yes	Yes
	8	Chinese	Ground	490	LPG	Local (K)	Mixed	Taegu (C)	Yes	Yes	Yes
	9	Western	Ground	190	LNG	Local (K)	Mixed	Taegu (R)	Yes	Yes	Yes
	10	Japanese	Ground	150	LPG	Local (K)	Mixed	Taegu (C)	Yes	Yes	Yes
	11	Korean	Ground	160	LPG	Local (K)	Mixed	Taegu (C)	Yes	Yes	Yes
	12	Korean	Ground	210	LPG, charcoal	Local (K)	Mixed	Taegu (R)	Yes	Yes	Yes

^a Fuel type: K—kerosene; B—briquet; L—light oil.^b Area: R—residential; C—commercial.

occupied by non-smokers. Two samples (each taken over a 2 h period) were collected, one when the evening meal was being prepared (18:00–20:00) and the other when at least one person was present (10:00–12:00). The ambient air samples were collected at street level at the same time as the indoor sampling occurred.

A non-smoking policy had been adopted at four of the 12 offices included in this study. Samples were taken during the morning (10:00–12:00) and in the afternoon (14:00–16:00), indoors and outdoors, at each office. In offices that did not have a non-smoking policy, active smoking occurred during sampling within the same room. The location of the building air intake was the determining factor so far as outdoor siting of the sampling equipment at the offices was concerned. If the main air intake was located on the roof, the outdoor sampling was conducted at that location; otherwise, sampling occurred at street level.

The restaurants selected for this study were typically Korean in character rather than international or of the "fast-food" variety. In each city, an assortment of restaurants of varying sizes and utilising a variety of cooking methods were studied. Samples were collected during lunchtime (12:00–14:00) and evenings (18:00–20:00). The indoor sampling equipment was positioned centrally within the main dining area and the ambient sampling was conducted at street level.

Unlike the other pollutants, NO_2 samples were collected in this study by a passive sampling method using filter badges (Advantec, Toyo Roshi Kaisha Inc., Japan). The badges were exposed for 24 h at each site, being attached to the wall closest to the sampling equipment in both indoor and outdoor environments. This extension to the sampling period was necessary to obtain adequate detection limits. Consequently, the number of samples for NO_2 collected in this study was only half those of other pollutants.

Sampling and analytical methods

VOCs. VOCs were collected by drawing air through a stainless steel sampling tube (1/4" × 9 cm, Perkin Elmer Ltd, U.K.) containing 300 mg of Carbotrap (60/80) using a personal air sampling pump at a flow rate of 50 ml min⁻¹ for a period of 2 h. Analyses of VOCs were performed by automatic thermal desorption coupled with capillary gas chromatography using a Perkin-Elmer ATD400 and an AutoSystem GC fitted with FID. The analytical column was an SGE BP1 capillary column (0.32 mm, 50 m, 1 µm). The column oven temperature programme was initially 40°C for 10 min, increased at a rate of 5°C min⁻¹ up to 200°C, and then maintained for 5 min at 200°C. The FID temperature was set to 300°C. The helium carrier gas flow rate was 2 ml min⁻¹ and the outlet split flow of the ATD400 was 10 ml min⁻¹. Before sampling, each tube was conditioned at 250, 300 and 330°C for 30 min consecutively in the helium carrier gas flow. A total of nine target VOCs were determined, including benzene, toluene, ethylbenzene, *o*-, *m*-, *p*-xylenes, styrene, 1,3,4-trimethylbenzene and 1,2,4-trimethylbenzene. Calibration was conducted by spiking clean adsorbent tubes with five levels (40, 80, 160, 400 and 800 µg ml⁻¹) of standard solutions (Supelco VOCs Mix2 and Mix7) and individual VOC stock solutions. A packed column injector was used to introduce the standard mixture into the flow of helium which carries the vaporised standards onto the tubes, and two tubes were connected in series to ensure that no breakthrough occurred.

CO, CO₂ and NO₂. Sampling for CO and CO₂ was performed for 2 h using 10 l Tedlar air sampling bags (SKC Inc., U.K.) at a flow rate of 50 ml min⁻¹. CO was determined using NDIR instrumentation (CO11M, Environment SA, France), while CO₂ was measured by gas chromatography. The GC (AutoSystem GC, Perkin Elmer Co., U.S.A.) was equipped with FID, a methaniser (Zr/Ni catalytic reactor, 350°C) and a six-port sampling valve. This enabled direct

injection from the Tedlar bags into the stainless steel analytical column (6' × 1/8") packed with Chromosorb 102 (60/80). The column was held at 40°C throughout the analysis and flushed with helium gas at a flow rate of 30 ml min⁻¹. Calibrations were undertaken using a standard gas mixture (CO 38.4 ppm, CO₂ 801 ppm in N₂) obtained from the Korean Institute of Standards and Science. NO₂ was measured by a passive filter badge method (Yanagisawa and Nishimura, 1980). The analytical method relies upon colour development with an azodye-forming reagent and subsequent analysis for UV absorbance at 545 nm.

RSP. RSP sampling was undertaken by drawing air through an impactor (Biral UK, Part No. 8508-035) with a cut-off 3.5 µm in diameter and a fluoropore membrane filter (FALP 02500, 1.0 µm pore, Millipore Co., U.S.A.) held in a 25 mm aluminium filter holder, using a personal pump operating at 2 l min⁻¹. Each filter was conditioned at 50% relative humidity for 24 h before and after sampling. Pre- and post-weighings (five replicates of each) were carried out using an electronic microbalance (Model M3P, Sartorius Inc., Germany). In order to obtain good precision, static charges were eliminated using a non-radioactive, ionising air blower (IB8, Amersham International, U.K.). The weight changes of blank field samples were evaluated to estimate the lower detection limit for RSP. Results indicated that the differences between blanks was rarely greater than 2 µg. Thus, a weight increase of at least 6 µg was required as a positive measurement, giving an RSP detection limit of 25 µg m⁻³ for a 2 h sample.

UVP. The particle-laden filter was extracted ultrasonically for 30 min in 2 ml of methanol. An aliquot of the extract was injected into a columnless high-performance liquid chromatograph (HPLC) system, and the UV absorbance was measured at 325 nm (Ogden *et al.*, 1990). The HPLC system consisted of a Rheodyne 7125 injector with a 100 µl sample loop, a Shimadzu SCL6B system controller, a SD6AV UV detector and a CR4A data integrator. An empty stainless steel tube (5 m × 0.33 mm) placed in a 30°C column oven was used to increase eluent retention times, with methanol as the mobile phase (1 ml min⁻¹). 2,2',4,4'-Tetrahydroxybenzophenone (THBP, 97% Aldrich) was used as a surrogate standard for ETS (Phillips *et al.*, 1994).

FPM. Determination of FPM was carried out simultaneously with the UVP determination on the same sample extract. Total sample fluorescence was measured at excitation 300 nm and emission 420 nm using a Shimadzu RF551 fluorescence detector connected in series with the UV detector. Scopoletin (95% Aldrich) was used as the surrogate standard for ETS (Ogden *et al.*, 1990).

SolPM. The solanesol content of the methanol extract was determined by reverse-phase HPLC using methanol as the mobile phase at a flow rate of 2 ml min⁻¹, a Shimadzu CLC-ODS column (15 cm × 4.6 mm) fitted with a C₁₈ guard column (5 cm × 4.6 mm) and UV detection at 210 nm (Ogden *et al.*, 1990). The solanesol standard (95% Sigma) was used without purification.

Nicotine. Nicotine was collected for 2 h by drawing air at a flow rate of 1 l min⁻¹ through glass tubes containing XAD-4 resin (SKC Inc., U.K.). After collection, the front and back sections of the resin were extracted in 1 ml of ethyl acetate, which contained 0.01% (v/v) triethylamine to prevent adherence to glassware, and 50 µl of a quinoline solution was added as an internal standard (Ogden *et al.*, 1989). Nicotine analyses were performed using a Hewlett Packard 5890II GC equipped with an autosampler, a nitrogen-specific detector and a DB5 megabore column (0.53 mm, 30 m, 1.5 mm). The column temperature was programmed from 150°C to 180°C at 5°C min⁻¹, while temperatures for the injector and detector were 250 and 260°C, respectively. The GC was operated in split injection mode with a split ratio of 5:1. The lower detection limit for nicotine was determined according to published guidelines (ACS Committee, 1980). This specifies a limit of three times the standard deviation

Table 2. Lower detection limits and precisions of the analytical methods

Air quality parameter	Lower detection limit (LDL)	No. of data below LDL (% of total)	Precision ^a
Benzene	0.2 $\mu\text{g m}^{-3}$	11 (3.8%)	2.5%
Toluene	0.2 $\mu\text{g m}^{-3}$	2 (0.7%)	4.5%
Ethylbenzene	0.2 $\mu\text{g m}^{-3}$	1 (0.3%)	5.0%
<i>m</i> + <i>p</i> -Xylene	0.5 $\mu\text{g m}^{-3}$	0 (0%)	4.3%
Styrene	0.3 $\mu\text{g m}^{-3}$	0 (0%)	4.2%
<i>o</i> -Xylene	0.3 $\mu\text{g m}^{-3}$	0 (0%)	4.3%
1,3,5-TMB ^b	0.2 $\mu\text{g m}^{-3}$	6 (2.2%)	3.9%
1,2,4-TMB	0.2 $\mu\text{g m}^{-3}$	1 (0.3%)	4.3%
RSP	25 $\mu\text{g m}^{-3}$	14 (4.9%)	0.1%
CO	0.1 ppm	0 (0%)	2.5%
CO ₂	1 ppm	0 (0%)	3.5%
NO ₂	4 ppb	0 (0%)	18.5%
Nicotine	0.6 $\mu\text{g m}^{-3}$	159 (55.2%)	4.5%
UVPm	1.5 $\mu\text{g m}^{-3}$	0 (0%)	2.1%
FPM	0.2 $\mu\text{g m}^{-3}$	11 (3.8%)	3.6%
SolPM	2.0 $\mu\text{g m}^{-3}$	206 (71.5%)	5.2%

^a Precision was expressed as a relative standard deviation of at least ten side-by-side analyses of a standard sample.

^b TMB: trimethylbenzene.

above the mean value of a series of field blanks. Under the sampling conditions used in this study, this resulted in a detection limit of 0.6 $\mu\text{g m}^{-3}$.

Miscellaneous data. Temperature and relative humidity were measured every 30 min indoors and outdoors. For each environment surveyed, various items of information about the sites were noted using a questionnaire. Questions included the ventilation type, size of the site, numbers of cigarettes smoked, heating type and other potential indoor pollution sources.

Detection limits and data quality control. Lower detection limits and precisions for the methods used in this study are presented in Table 2. In general, detection limits were determined by taking account of results both from the field blanks and the signal to noise (S/N) ratio of the analysed samples and calibration standards. A S/N ratio of three was required for a positive chromatographic response, whilst for CO the instrumentation detection limit was used. In many cases, particularly for SolPM and nicotine, levels were found to be below the lower detection limit. This raised the questions of how to deal with these data in the statistical analysis. As a reasonable compromise, a value of one half the detection limit was used for these data (Hornung and Reed, 1990), as is common in other studies of this type (Kirk *et al.*, 1988; Chan *et al.*, 1991; Daisey *et al.*, 1994; Phillips *et al.*, 1994).

RESULTS AND DISCUSSION

Indoor and ambient concentrations

Summary statistics for the concentrations of all the measured pollutants in indoor and outdoor air are given in Tables 3–5. The sample size for each of the analytes in each category is 48, apart from NO₂ for which the size is 24. As shown in Tables 3–5, arithmetic means are higher than the medians, indicating that the concentration data are log-normally distributed. This distribution is typical of indoor air quality data and air pollutants in general (Cohen *et al.*, 1989).

Ratios of median indoor concentrations to outdoor ones (I/O ratios) for each analyte are also included in Tables 3–5, together with the Wilcoxon rank sum test *p* values for determining statistically significant differences between indoor and outdoor levels. The Wilcoxon test was used because the data are not normally distributed (Heavner *et al.*, 1994).

The data presented in Tables 3–5 are summary data for the two cities monitored and contain data collected at different times of the day. In order to examine temporal variations in indoor and ambient air quality during the day, ratios of median concentrations of the first to the second sample groups were calculated for each pollutant. No statistically significant differences were found between the two groups of data, except for one indoor RSP data set for restaurants. This demonstrates that at the sampling frequency and duration used in this study, morning, afternoon, and evening measurements are broadly similar. Hence the two groups of samples can be regarded as having been selected from the same population. Due to the longer sampling time (24 h), no information could be determined concerning the temporal variation of NO₂.

Locational differences between the samples collected in Seoul and Taegu were investigated in the same manner. Wilcoxon rank sum tests of significance showed that most of the differences between the two cities were not significant for either indoor or outdoor measurements. Although some of the VOCs data exhibited significant differences (*o*-xylene, 1,3,5- and 1,2,4-trimethylbenzenes), it is not clear if this was due to real differences or to the small data populations affecting the Wilcoxon test's response. Similarly, locational differences between residential and commercial areas were investigated. Concentrations of CO, CO₂ and most of the VOCs were found to be significantly higher in the commercial districts. This may indicate the influence of increased vehicle emissions, on both indoor and outdoor air, which might be expected in commercial districts.

It can be seen from Tables 3–5 that the indoor and ambient levels of the pollutants measured in this study varied widely between the three types of environments studied. When comparing median values, restaurants had higher indoor levels of most pollutants than homes or offices, indicating the effect of human activity on indoor air quality. Concentrations of RSP, CO and NO₂ in restaurants were not only higher but were more variable than those measured in homes and offices, as indicated by the higher standard deviations and maximum and minimum values for these pollutants. However, VOC levels were found to be more variable in offices than in restaurants and homes. In several instances, CO₂ levels were determined to be greater than 1000 ppm. In buildings where there is considerable human occupancy, levels this high are typically taken to be indicative of poor ventilation. Together, these results imply that under some circumstances indoor sources can have a considerable

Table 3. Summary statistics for indoor and outdoor concentrations (in $\mu\text{g m}^{-3}$) of target pollutants in homes

Air quality parameter	Indoor				Outdoor				I/O ratio ^a	WRST ^b <i>p</i>
	Median	Mean	S.D.	Range	Median	Mean	S.D.	Range		
RSP	91	100	58	12-244	68	80	58	12-271	1.3	0.049 ^d
CO (ppm)	1.8	2.1	1.5	0.1-6.2	1.6	2.1	1.6	0.2-7.0	1.1	0.717
CO ₂ (ppm)	685	790	494	369-3634	494	577	366	357-2881	1.4	< 0.001 ^d
NO ₂ (ppb)	24	33	22	9-96	29	32	14	12-67	0.8	0.510
Benzene	7.9	8.2	7.9	0.1-27.2	6.9	9.4	10.9	0.1-47.2	1.1	0.823
Toluene	31.0	42.3	47.2	0.6-202.4	27.4	42.6	54.4	0.1-222.1	1.1	0.742
Ethylbenzene	4.0	5.1	4.1	0.6-17.0	3.9	5.1	5.0	0.1-23.1	1.0	0.753
<i>m</i> + <i>p</i> -Xylene	11.6	14.4	11.2	2.0-57.4	10.6	14.8	14.1	0.8-59.9	1.1	0.725
Styrene	3.7	3.9	1.8	0.6-8.3	3.3	3.9	2.9	0.3-15.6	1.1	0.448
<i>o</i> -Xylene	7.0	9.1	7.0	1.3-29.1	5.5	8.6	8.2	0.7-36.7	1.3	0.558
1,3,5-TMB ^c	3.1	4.1	4.6	0.3-25.0	2.5	4.4	8.8	0.1-59.4	1.2	0.281
1,2,4-TMB	6.8	9.5	11.7	1.0-71.4	5.8	9.9	17.9	0.1-108	1.2	0.225
Nicotine	0.6	1.8	2.5	0.3-9.5	0.3	0.3	0.1	0.3-0.7	2.0	< 0.001 ^d
UVPM	13.2	19.5	23.1	3.3-146	6.4	9.5	8.1	1.6-42.0	2.1	< 0.001 ^d
FPM	6.0	9.3	11.9	0.6-77.9	3.4	4.2	3.8	0.1-15.7	1.8	< 0.001 ^d
SolPM	1.0	7.0	19.9	1.0-131	1.0	1.5	2.1	1.0-12.2	1.0	0.003 ^d

^a Ratio of median concentrations indoors to outdoors.^b WRST *p*: Wilcoxon rank sum test significance.^c TMB: Trimethylbenzene.^d Indicates that indoor and outdoor groups are different at a significant level of 0.05 by the Wilcoxon rank sum test.

Table 4. Summary statistics for indoor and outdoor concentrations (in $\mu\text{g m}^{-3}$) of target pollutants in offices

Air quality parameter	Indoor				Outdoor				I/O ratio ^a	WRST ^b <i>p</i>
	Median	Mean	S.D.	Range	Median	Mean	S.D.	Range		
RSP	92	99	68	12-392	70	78	44	12-197	1.3	0.046 ^d
CO (ppm)	1.9	2.4	2.0	0.4-6.8	1.9	2.2	1.6	0.5-6.1	1.0	0.866
CO ₂ (ppm)	770	952	752	372-4016	439	569	375	347-2287	1.8	< 0.001 ^d
NO ₂ (ppb)	19	22	10	9-58	29	31	13	10-73	0.7	< 0.001 ^d
Benzene	6.2	12.6	19.1	0.1-108.8	3.8	8.2	10.3	0.1-49.9	1.6	0.324
Toluene	34.4	80.4	95.3	0.8-340.8	14.6	50.4	65.8	0.5-270.6	2.4	0.205
Ethylbenzene	6.2	7.6	8.9	0.6-44.8	3.7	5.5	6.1	0.6-28.1	1.7	0.187
<i>m</i> + <i>p</i> -Xylene	20.2	23.4	26.8	2.0-136.8	12	16.2	17.6	2.2-96.4	1.7	0.159
Styrene	4.1	5.0	4.3	1.6-27.4	3.2	4.0	2.4	1.6-11.8	1.3	0.101
<i>o</i> -Xylene	10.4	14.5	16.9	1.1-75.2	6.1	9.0	9.6	1.1-48.7	1.7	0.095
1,3,5-TMB ^c	2.3	6.4	14.0	0.1-76.4	2.5	2.9	2.4	0.1-11.7	0.9	0.577
1,2,4-TMB	6.4	14.6	29.4	0.5-155.0	5.1	6.5	5.4	0.6-27.0	1.3	0.130
Nicotine	1.1	2.5	3.9	0.3-22.7	0.3	0.4	0.1	0.3-0.7	3.7	< 0.001 ^d
UVPM	12.6	19.9	23.4	3.2-133	8.3	10.8	8.5	2.0-33.6	1.5	0.015 ^d
FPM	6.6	8.7	9.0	0.1-48.2	3.3	4.7	4.4	0.1-19.4	2.0	0.014 ^d
SolPM	3.0	14.2	28.7	1.0-139	1.0	1.0	0	1.0-1.0	3.0	< 0.001 ^d

^a Ratio of median concentrations indoors to outdoors.^b WRST_p: Wilcoxon rank sum test significance.^c TMB: trimethylbenzene.^d Indicates that indoor and outdoor groups are different at a significant level of 0.05 by the Wilcoxon rank sum test.

Table 5. Summary statistics for indoor and outdoor concentrations (in $\mu\text{g m}^{-3}$) of target pollutants in restaurants

Air quality parameter	Indoor				Outdoor				I/O ratio ^a	WRST ^b <i>p</i>
	Median	Mean	S.D.	Range	Median	Mean	S.D.	Range		
RSP	159	171	101	33-475	67	72	40	12-172	2.4	< 0.001 ^d
CO (ppm)	4.4	11.3	16.4	0.7-89.9	2.2	2.4	1.6	0.4-6.6	2.0	< 0.001 ^d
CO ₂ (ppm)	1034	1081	488	432-2220	506	522	120	372-963	2.0	< 0.001 ^d
NO ₂ (ppb)	56	58	22	21-105	40	42	14	19-77	1.4	< 0.001 ^d
Benzene	6.9	12.0	12.8	0.1-51.7	7.6	8.0	8.2	0.3-31.1	0.9	0.229
Toluene	42.8	52.0	55.4	0.1-234.1	30.0	49.5	55.2	1.3-212.6	1.4	0.719
Ethylbenzene	6.6	8.2	6.5	0.1-33.6	4.3	7.4	12.6	1.0-87.7	1.5	0.046 ^d
<i>m</i> + <i>p</i> -Xylene	19.1	22.4	14.6	0.2-71.2	14.1	20.8	19.6	4.4-99.9	1.4	0.131
Styrene	3.8	4.1	2.0	0.1-9.3	3.1	3.4	1.5	1.1-7.2	1.2	0.052
<i>o</i> -Xylene	11.3	13.0	7.6	0.1-36.6	8.1	11.2	9.8	3.1-50.2	1.4	0.027 ^d
1,3,5-TMB ^c	4.1	5.1	4.3	0.1-24.3	3.0	3.8	3.4	0.9-19.2	1.4	0.051
1,2,4-TMB	8.6	10.9	8.6	0.1-49.1	6.5	9.3	8.1	2.7-40.0	1.3	0.146
Nicotine	3.0	4.8	5.6	0.3-28.9	0.3	0.4	0.2	0.3-1.0	10.0	< 0.001 ^d
UVP	32.9	45.6	39.2	3.6-159	5.0	7.5	6.6	1.5-40.9	6.6	< 0.001 ^d
FPM	17.4	22.5	19.1	1.3-78.3	2.7	3.5	3.3	0.1-18.0	6.4	< 0.001 ^d
SolPM	15.2	25.4	32.0	1.0-124	1.0	1.2	0.8	1.0-5.2	15.2	< 0.001 ^d

^a Ratio of median concentrations indoors to outdoors.^b WRST *p*: Wilcoxon rank sum test significance.^c TMB: trimethylbenzene.^d Indicates that indoor and outdoor groups are different at a significant level of 0.05 by the Wilcoxon rank sum test.

impact on the local indoor concentrations of specific pollutants and that the sources of VOCs in various indoor environments might be significantly different from those of RSP, CO, CO₂ and NO₂.

A clear pattern is also apparent in Tables 3–5 of the indoor levels of target pollutants being higher than those outdoors (I/O ratios > 1), particularly in restaurants. Thus, whilst the ambient air may have a predominating influence on the levels of most indoor air constituents, activities and materials found indoors were shown in this study to contribute significantly to indoor pollution in many instances.

VOC concentrations. A relatively large number of VOC studies are available in the literature for both ambient (Singh *et al.*, 1992; Evans *et al.*, 1992; Field *et al.*, 1992; Shah and Singh, 1988; Edgerton *et al.*, 1989) and indoor air (Hartwell *et al.*, 1987; Holcomb and Seabrook, 1995; Daisey *et al.*, 1994; Kostianen, 1995). The indoor and ambient VOC levels measured in this study are generally consistent with previous research. Despite the fact that a number of VOCs have been identified as important cancer risk factors (Tancrede *et al.*, 1987; Feron *et al.*, 1991; U.K. DOE, 1994) and are implicated in the formation of photochemical oxidants (Singh *et al.*, 1992), they have not been routinely measured in Korea and no national ambient air quality standards have been established for VOCs. The VOC analyses made during this study are to our knowledge part of the first extensive monitoring programme for VOCs in the non-occupational environment in Korea.

A very wide range of VOC levels were documented in this study. For example, benzene levels ranged from undetectable ($< 0.2 \mu\text{g m}^{-3}$) up to $108.8 \mu\text{g m}^{-3}$ (Tables 3–5). Although the level of individual VOCs varied between sites, their relative abundances (i.e. the ratio of each VOC to the total of the nine VOCs studied) were generally the same. Toluene and *m* + *p*-xylenes were the most abundant compounds, followed by *o*-xylene and 1,2,4-trimethylbenzene. Although median indoor/outdoor ratios indicate a generally increasing level of VOCs indoors when compared to outdoors, no statistically significant differences (at a level of 0.05) were found between indoor and outdoor VOC levels in homes and offices. This indicates that the major sources of all the VOCs monitored in this study were outdoors. Indoor levels of ethylbenzene and *o*-xylene at restaurants were, however, found to be significantly different, implying an additional indoor source for those pollutants.

It has been suggested that the relative ratios of different aromatic compounds may be useful in identifying the sources of VOCs in urban areas (Evans *et al.*, 1992). The ratios of benzene to toluene measured in a variety of studies in different parts of the world usually fall into the range 1:2 to 1:4 for ambient air (Evans *et al.*, 1992; Chan *et al.*, 1991; Bevan *et al.*, 1991). These are close to the levels reported for vehicle emissions (Scheff *et al.*, 1989; Sweet and Vermette, 1991). In this study, ambient levels of

benzene to toluene ratios were fairly constant at 1:4, which is similar to previous studies. The similarity of the measured ratios in Korea to those found in other regions imply that vehicle emissions are likely to contribute to the majority of VOCs monitored outdoors in the two Korean cities. The variation in the reported ratios and those measured in Korea can perhaps be explained by differences in fuel composition and vehicle types in the different regions and countries.

Indoor benzene to toluene ratios are usually more variable than for ambient air due to the wide variety of potential indoor sources and the complexity of the indoor environment (Holcomb and Seabrook, 1995; Daisey *et al.*, 1994; Fellin and Otson, 1994; Crump and Madany, 1993). In this study, benzene to toluene ratios of 1:4 were found for homes and 1:6 for offices and restaurants. These results suggest that offices and restaurants are influenced by additional indoor sources (possibly kerosene heaters and charcoal barbecues) where homes are more strongly affected by the migration of VOCs from ambient air.

Seasonal variations

In order to investigate the effect of seasonality on indoor and ambient air quality, winter to summer median ratios were calculated, and the results are shown in Table 6. The Wilcoxon rank sum test was also carried out to test the significance of the two sets of winter and summer data. It was apparent that there is a general pattern of increasing levels from summer to winter, and similarly from outdoor to indoor air, for nearly all of the substances measured in this study.

Seasonal differences were noted for pollutants related to fossil fuel combustion (CO, RSP, CO₂ and VOCs), most probably due to the effects of increased fuel consumption, poor operation of catalytic converters and meteorological factors. Indoor levels of CO in homes and offices generally were low in both summer and winter. In Korea, coal briquettes (Yeontan) traditionally have been used for heating in the winter months. These are either burnt outside the house or inside the kitchen, with the chimney passing underneath the rooms to provide under-floor heating. As a result of this system, CO poisoning was once very common in Korea as chimney gases seeped into rooms during stagnant winter nights. Recently, however, the use of coal briquettes in the residential sector has declined dramatically following governmental initiatives to substitute "dirty" fuels with cleaner alternatives (Baek, 1993). The results of this study indicate that such schemes have been reasonably successful since indoor CO levels were not significantly different from those found outdoors.

Two very high CO₂ measurements were made during the winter inside office #11 (4016 and 3907 ppm — about eight times the outdoor level), along with high RSP levels. Two unvented kerosene stoves were used in this office during the sampling period, which probably explains these results. Unusually high CO₂ concentrations were also measured indoors and outdoors

Table 6. Seasonal differences of median concentrations of target pollutants in indoor and outdoor air of homes, offices, and restaurants

Air quality parameter	Ratio of median concentrations in winter to summer					
	Home		Office		Restaurant	
	In	Out	In	Out	In	Out
RSP	1.7 ^a	2.4 ^a	1.6 ^a	1.9 ^a	1.7 ^a	1.1
CO	4.3 ^a	3.8 ^a	5.9 ^a	4.4 ^a	4.6 ^a	3.1 ^a
CO ₂	1.5 ^a	1.1	2.0 ^a	1.4 ^a	2.1 ^a	1.1
NO ₂	1.4	0.9	1.0	0.8 ^a	1.6 ^a	0.6 ^a
Benzene	8.8 ^a	9.8 ^a	18.8 ^a	14.4 ^a	14.9 ^a	8.1 ^a
Toluene	9.2 ^a	8.6 ^a	27.1 ^a	15.1 ^a	9.3 ^a	6.8 ^a
Ethylbenzene	4.9 ^a	3.8 ^a	5.4 ^a	3.6 ^a	1.4 ^a	1.2 ^a
<i>m</i> + <i>p</i> -Xylene	3.6 ^a	3.0 ^a	4.8 ^a	2.9 ^a	1.3 ^a	1.0
Styrene	1.2	1.3 ^a	1.7 ^a	1.0	1.2 ^a	1.2
<i>o</i> -Xylene	3.1 ^a	2.7 ^a	3.9 ^a	2.2 ^a	1.3 ^a	1.0
1,3,5-TMB	3.8 ^a	3.5 ^a	5.7 ^a	3.3 ^a	2.2 ^a	1.5 ^a
1,2,4-TMB	2.5 ^a	2.4 ^a	3.9 ^a	1.7 ^a	1.3 ^a	1.0
Sample size	24	24	24	24	24	24

^a Denotes that winter and summer data groups are significantly different at a level of 0.05.

at home #5, which may have been associated with the burning of refuse near that site.

A notable exception to the generally higher winter pollutant levels was found for NO₂, particularly for outdoor samples, where lower median levels were measured in the winter (Table 6). Ambient levels of NO₂ measured at fixed monitoring sites, that were not a part of this study, indicate that average NO₂ concentrations are 1.5–2 times higher in the winter than in the summer in Seoul and Teagu, which is contradictory to our findings. Such differences call into question the sensitivity of the filter badge sampling method that was utilised in this study for NO₂. The badges were exposed for 24 h in this study, giving a reported precision of 20% for wind velocities in the range 0.15–4 m s⁻¹ and 40–80% of relative humidity (Yanagisawa and Nishimura, 1980). This method was originally developed for monitoring personal exposure indoors. The uncertainty associated with the method may be increased when the badge is exposed outside buildings in conditions of higher wind velocities, which can occur during the winter months in Korea. Caution is therefore warranted when assessing the outdoor NO₂ winter results contained in Tables 6 and 7.

There was a marked seasonal variation in VOC levels both indoors and outdoors. Median ambient levels of benzene were 8–15 times higher during the winter than during the summer (Table 6). Other VOC levels also were consistently higher during the winter months. In contrast to CO and RSP results, maximum VOC levels were all found in offices, especially offices #2 and #8. Unvented kerosene stoves were used during the winter in both offices, which undoubtedly explains the results from those locations. Interestingly, smoking was not allowed in either office and cannot therefore have been responsible for the elevated VOC levels.

Increased VOC levels generally are expected during the winter (Rasmussen and Khalil, 1983; Tille *et al.*, 1985; Singh *et al.*, 1992; Field *et al.*, 1992). The seasonal variation in VOC levels can be influenced by a number of factors, including source variation, fuel consumption, chemical reactivity, meteorological changes, and the location and time of sampling. Warmer temperatures in the summer will obviously increase the evaporation of VOCs from vehicle fuel tanks. But the decay or removal of VOCs through photochemical reactions will be more significant during the summer than during the winter (Dann and Wang, 1995). In addition, ambient VOC levels tend to increase due to air stagnation (Singh *et al.*, 1992). Finally, tailpipe emissions of benzene and other VOCs from motor vehicles are sensitive to changes in the aromatic content of fuel and catalytic control systems, which are both influenced by climatological factors (Black *et al.*, 1980; Perry and Gee, 1994).

The number of vehicles in Korea has risen from 0.8 million in 1984 to 8 million in 1995, a 10-fold increase in about 10 years. As a consequence, the control of vehicle emissions has become an important part of government policy in Korea. From July 1987, all new cars have been required to be equipped with three-way catalytic converters and to use unleaded fuel. This policy was extended in 1988 to include all passenger cars. Catalytic converters have recognised limitations, the most important of which is the warm-up period required for the catalyst to reach an effective operating temperature (Perry and Gee, 1994). Since the majority of car journeys cover only a few kilometres, the catalyst will be ineffective over much of the journey. It is well known that cold-start VOC emissions account for a large proportion of the total VOC emissions from motor vehicles, particularly during the winter. Problems also exist with inadequate maintenance and catalyst poisoning, which further reduce the

effectiveness of catalytic converters. The increased winter VOC levels observed (Table 6) imply that catalysts do not remove sufficient VOCs from exhaust to prevent accumulation under stagnant winter conditions in Korea. These vehicle-related factors, together with the extensive use of fossil fuel heating systems under stagnant air conditions, can explain much of the dramatic increase in VOC levels observed in this study during the winter months, both indoors and outdoors.

Correlations between indoor and outdoor air quality

A number of studies have demonstrated that outdoor air quality can have a significant impact on indoor air (Yocom, 1982; Daisey *et al.*, 1994; Perry and Gee, 1994). To investigate relationships between indoor and outdoor air quality, correlation analyses were performed on the indoor vs outdoor concentrations of each pollutant in the three environments studied. For those pollutants displaying significant correlations between indoor and outdoor levels, a source relationship is implied. In this study, correlation coefficients were calculated for the summer and winter data sets separately (Table 7), since the response of indoor air to any changes in outdoor air quality depends upon a number of seasonal factors such as heating facilities, ventilation, building permeability and pollutant stability.

It is important to note that the sample sizes used for the correlation analyses in this study were relatively small ($n = 24$), and there is a possibility that the correlation coefficients were significantly affected by one or two extremely high levels of data. For these reasons, interpretation of the correlation results in Table 7 should be viewed as suggestive rather than definitive.

In general, very good correlations were identified between indoor and outdoor air quality in homes during both summer and winter periods. Correlations for the offices and restaurants were more variable for

the two seasons, and for each pollutant. When taking into account the summer data set only, there are consistently good correlations between indoor and outdoor VOC concentrations for both homes and offices. This suggests that during summer there is a strong relationship between outdoor VOC sources, mainly vehicle emissions, and indoor VOC concentrations in homes and offices. For the restaurants, however, it is not clear that the contribution of vehicle sources to indoor levels of VOCs is as important as for homes and offices. Several potential VOC sources are present inside restaurants even during the summer season and the very poor correlations obtained for RSP and CO also indicate the importance of such sources.

The results for the winter data sets are more difficult to explain because the indoor/outdoor relationships were complicated by other factors that affect indoor air quality during the cold season. As low correlations were observed for CO₂, it is likely that the lower correlations observed for the other pollutants were largely due to limited ventilation during the winter.

Other factors affecting indoor air quality

The trend of increased indoor pollution during the winter months when compared with the summer months implies that several factors influence indoor air quality during the winter in addition to outdoor air and meteorological factors. Such factors include indoor activities, duration of human occupancy and ventilation. Indoor activities that generate pollutants include the use of gas or kerosene stoves for heating and cooking, cleaning and the use of a variety of consumer products, including tobacco smoking. The density of human occupancy, with people tending to spend more time indoors in the winter than in the summer, combined with inadequate ventilation also can play important roles in determining air quality in the winter.

Table 7. Correlations between indoor and outdoor air quality

Air quality parameter	Summer			Winter		
	Home	Office	Restaurant	Home	Office	Restaurant
RSP	0.63 ^a	0.17	-0.01	0.76 ^a	0.25	0.02
CO	0.37	0.48 ^a	-0.04	0.72 ^a	0.34	0.32
CO ₂	0.96 ^a	0.71 ^a	0.77 ^a	0.18	-0.12	0.41
NO ₂	0.79 ^a	0.79 ^a	0.52 ^a	0.42	0.23	0.53 ^a
Benzene	0.91 ^a	0.56 ^a	0.14	0.72 ^a	0.70 ^a	0.27
Toluene	0.88 ^a	0.55 ^a	0.75 ^a	0.85 ^a	0.30	0.85 ^a
Ethylbenzene	0.93 ^a	0.82 ^a	0.33	0.72 ^a	0.33	0.84 ^a
<i>m</i> + <i>p</i> -Xylene	0.94 ^a	0.85 ^a	0.89 ^a	0.73 ^a	0.52 ^a	0.67 ^a
Styrene	0.78 ^a	0.60 ^a	0.72 ^a	0.51 ^a	-0.04	0.37
<i>o</i> -Xylene	0.83 ^a	0.92 ^a	0.88 ^a	0.72 ^a	0.34	0.70 ^a
1,3,5-TMB	0.99 ^a	0.70 ^a	0.75 ^a	0.47 ^a	0.06	0.40
1,2,4-TMB	0.99 ^a	0.65 ^a	0.70 ^a	0.46 ^a	0.11	0.36
Sample size	24	24	24	24	24	24

^a $p < 0.01$.

Environmental tobacco smoke (ETS). To assess the impact of smoking on the indoor air quality, nicotine and other ETS-related components (UVPM, FPM, SolPM) were measured together with the other pollutants. Summary statistics for these parameters are included in Tables 3–5. Figure 1 compares the median ratios for indoor to outdoor concentrations (I/O ratios) of air pollutants at smoking and non-smoking homes and offices (restaurants are not included as smoking occurred at all the sites studied). The I/O ratios were calculated by taking the ratio between indoor and outdoor measurements for each matched sample. In non-smoking homes and offices, I/O ratios were generally close to 1.0, indicating that the outdoor air was the overwhelmingly predominant indoor pollution source at those sites. In smoking homes and offices, however, I/O ratios for at least some pollutants were significantly greater than 1.0, implying additional indoor sources.

To investigate this further, correlation coefficients were calculated for the VOCs, CO, RSP and ETS parameters monitored in smoking indoor environments (Table 8). The number of cigarettes smoked during the sampling period was also included in the correlation analysis. It is clear that nicotine and the other ETS parameters (UVPM, FPM, SolPM, and the number of cigarettes smoked) were not strongly correlated with indoor VOC or CO levels measured in this study. This is strong evidence that smoking is not a major factor in determining VOC or CO levels in the indoor environment in Korea, although it does not rule out a contributory role. By contrast, a very strong correlation was generally found between the indoor and outdoor levels of vehicle-related pollutants (Table 7), including CO, benzene and the other VOCs, especially for homes and offices in the summer when there are fewer indoor pollution sources such as space heaters, as discussed previously. This supports

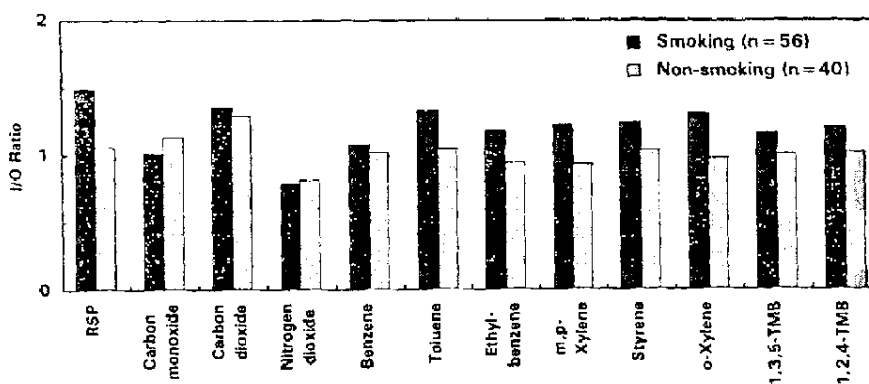


Fig. 1. Comparison of median ratios of indoor (I) and outdoor (O) levels of target pollutants in smoking and non-smoking homes and offices.

Table 8. Coefficients of correlation between ETS components and VOCs in smoking indoor environments ($n = 104$)

VOC	RSP	CO	Nicotine	UVPM	FPM	SolPM	No. of cigarettes
RSP	1.00						
CO	0.56 ^a	1.00					
Nicotine	0.42 ^a	0.11	1.00				
UVPM	0.74 ^a	0.14	0.52 ^a	1.00			
FPM	0.71 ^a	0.26 ^a	0.57 ^a	0.97 ^a	1.00		
SolPM	0.50 ^a	0.07	0.60 ^a	0.75 ^a	0.73 ^a	1.00	
No. of cigarettes	0.36 ^a	0.35 ^a	0.50 ^a	0.41 ^a	0.46 ^a	0.44 ^a	1.00
Benzene	0.44 ^a	0.52 ^a	0.23 ^a	0.16	0.16	0.22	0.16
Toluene	0.25 ^a	0.16	0.20	0.12	0.08	0.29 ^a	0.01
Ethylbenzene	0.25 ^a	0.13	0.10	0.12	0.11	0.13	-0.02
m + p-Xylene	0.31 ^a	0.14	0.11	0.11	0.11	0.13	-0.04
Styrene	0.27 ^a	0.20	0.14	0.04	0.05	0.09	0.12
o-Xylene	0.28 ^a	0.17	0.10	0.06	0.08	0.09	-0.04
1,3,5-TMB	0.34 ^a	0.17	0.17	0.06	0.09	0.09	-0.04
1,2,4-TMB	0.27 ^a	0.05	0.06	0.03	0.06	0.05	-0.10

^a $p < 0.01$.

the conclusion that vehicle emissions, migrating indoors from the ambient air, rather than ETS, are the most important factor influencing indoor VOC levels.

Although smoking appears to increase VOC levels in homes and offices (Fig. 1), the increase documented in this study was not statistically significant. Thus, in comparison to outdoor sources, ETS must be regarded as only a minor contributor to indoor levels of VOC and CO. At the same time, the I/O ratios for RSP found in this study were higher in smoking homes/offices than in non-smoking homes/offices (Fig. 1). Further, RSP levels indoors correlated with ETS components, including UVPM, FPM and to a lesser degree SolPM and nicotine (Table 8). This suggests that RSP levels are influenced by smoking to a greater degree than VOCs or CO levels.

The extent of the contribution of smoking to indoor RSP levels has become a subject of concern (Ogden *et al.*, 1990; Phillips *et al.*, 1994). To estimate the contribution of ETS to RSP levels, three particle-phase ETS markers were determined; UVPM, FPM and SolPM. These were used to calculate RSP equivalent concentrations using two sets of conversion factors available in the literature (Ogden *et al.*, 1990; Phillips *et al.*, 1994). The average contributions of ETS to RSP levels

were estimated by calculating the average concentrations of the three ETS markers as a percentage of the average RSP concentrations in each environment (homes, offices and restaurants). The data are presented in Table 9. The highest calculated ETS contribution to RSP was a level of 35% in restaurants based on UVPM levels calculated using the conversion factors of Ogden *et al.* (1990). However, because UVPM can emanate from any combustion source, UVPM measurements tend to produce an overestimate of smoking's contribution to RSP levels. It has been suggested that the FPM and SolPM methods are more specific for ETS particulate matter than the UVPM method (Ogden *et al.*, 1990). In this study, the mean FPM levels were approximately 50% of the UVPM values. The ETS contributions based upon FPM and SolPM fell within a range of 10–20% of the average RSP levels measured in smoking environments in both cities. Thus, these results indicate that ETS contributes, on average, no more than 20% of the total RSP load in the buildings monitored in this study.

Cooking and heating types. The use of kerosene stoves has long been suspected of being responsible for the most extreme levels of certain pollutants

Table 9. Estimated average contributions of ETS to RSP in indoor smoking environments

Site	RSP	Estimation was made using conversion factors determined by					
		Phillips <i>et al.</i> (1994)			Ogden <i>et al.</i> (1990)		
		UVPM	FPM	SolPM	UVPM	FPM	SolPM
Home (n = 26)	Mean ($\mu\text{g m}^{-3}$) % of RSP	110 22.7%	12 10.9%	12 10.9%	33 30.3%	18 16.4%	9 8.2%
Office (n = 30)	Mean ($\mu\text{g m}^{-3}$) % of RSP	96 25.0%	10 10.4%	21 21.9%	32 33.3%	15 15.6%	16 17.1%
Restaurant (n = 48)	Mean ($\mu\text{g m}^{-3}$) % of RSP	171 26.9%	23 13.5%	25 14.6%	60 35.1%	34 19.9%	20 11.6%

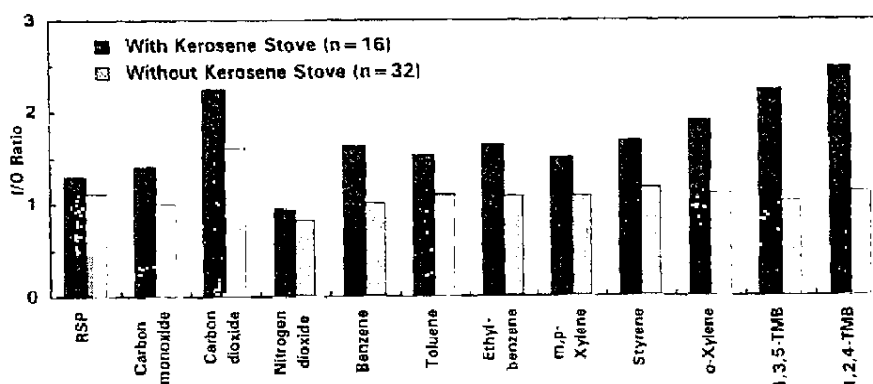


Fig. 2. Comparison of median ratios of indoor (I) and outdoor (O) levels of target pollutants in homes and offices with and without kerosene stoves during winter.

during the winter in Korea. In order to establish the impact of unvented kerosene stoves on indoor air in Korean homes and offices during the winter, the median indoor/outdoor ratios of the monitored pollutants were compared (Fig. 2). It is clear that the use of kerosene heaters greatly increases the level of almost all of the monitored pollutants. The levels of CO_2 and VOCs were particularly affected by the use of such stoves. The use of alternative fuels or heater designs should therefore be considered to help improve indoor air quality in Korea and in other countries such as Japan, where the use of portable kerosene heaters is also common (Kodama *et al.*, 1991; Kawamoto *et al.*, 1993).

In many Korean restaurants, barbecue-style cooking employing charcoal burners is widespread. The median ratios for indoor to outdoor levels were studied for different restaurants where gas only and gas and charcoal cooking were utilised (Fig. 3). The use of charcoal burners does not appear to influence indoor VOC levels. It is associated, however, with an increase in the indoor levels of RSP and CO. An

interesting feature is that extremely high levels of CO and RSP were found in the indoor air of particular restaurants. Indoor CO levels were as high as 90 ppm in restaurant #5 and 41 ppm in restaurant #1. RSP levels in restaurant #1 were $425 \mu\text{g m}^{-3}$ in the summer and $475 \mu\text{g m}^{-3}$ in the winter. These restaurants were both Korean barbecue restaurants using numerous charcoal burners upon which customers cook their own food. The use of such burners clearly can have a major impact on indoor air quality, particularly during the winter under conditions of reduced ventilation.

Air conditioning systems. Air conditioning can and often does have a positive impact on indoor air quality. This depends in part, however, upon the standard of maintenance and operation of the system being utilised in any given building. To investigate the effectiveness of the air conditioning systems installed in Korean homes and offices, indoor/outdoor ratios of the studied pollutants were calculated (Fig. 4). For most of the pollutants, buildings with air conditioning had low I/O ratios, implying that the majority of the

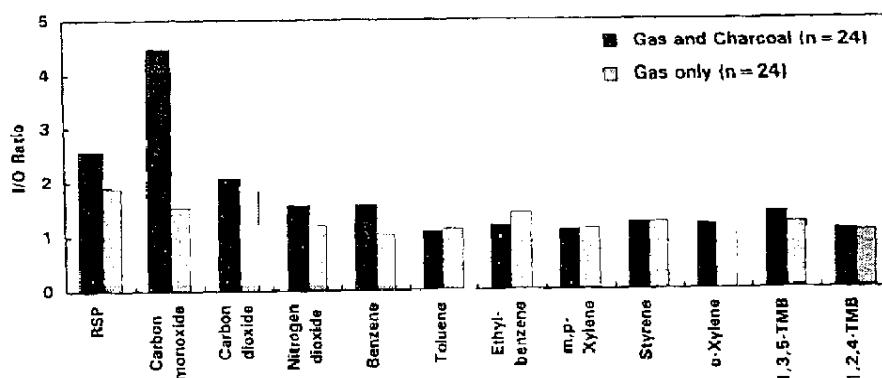


Fig. 3. Comparison of median ratios of indoor (I) and outdoor (O) levels of target pollutants in restaurants by cooking fuel type: gas only and gas with charcoal.

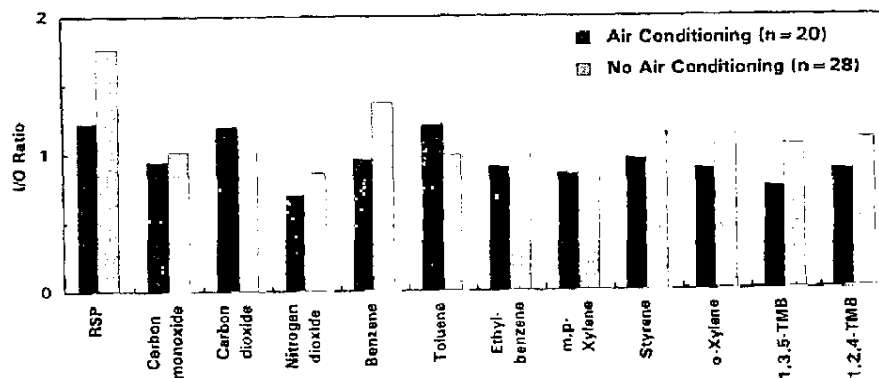


Fig. 4. Comparison of median ratios of indoor (I) and outdoor (O) levels of target pollutants in homes and offices with and without air conditioning during summer.

air conditioning systems installed in the buildings included in this study operated effectively. The greatest impact was observed for RSP, as would be expected due to the filtration devices used, and for benzene. However, although RSP levels tended to be reduced in the air-conditioned buildings, I/O ratios were still greater than 1.0, indicating that indoor levels were higher than outdoors. Toluene and CO₂ displayed the opposite effect, with higher I/O ratios being found in buildings with air conditioning. This may be due to air recirculation within the building, which would tend to build up CO₂ produced by occupants and also toluene if there were indoor toluene sources.

CONCLUSIONS

This study has confirmed the importance of ambient air quality in determining the quality of air indoors in two major Korean cities. Although the quality of the indoor air, as documented in this study, was generally poorer than the quality of the air outdoors, indoor air was strongly influenced by outdoor sources of pollution.

The majority of the VOCs measured in this study in both indoor and outdoor environments can be attributed to outdoor sources, motor vehicles in particular. Benzene concentrations were much higher during the winter months and were not significantly greater in smoking than in non-smoking sites. Heating and cooking practices, coupled with generally inadequate ventilation, were also shown to influence indoor air quality with respect to VOCs. These factors were particularly prevalent during the winter months.

ETS was found in this study to have been a minor contributor to total VOC levels as no statistically significant relationships were identified between ETS components and VOCs, whereas a very strong correlation was found between indoor and outdoor levels of VOC pollutants, usually associated with vehicles. This would suggest that vehicles are the major contributor to indoor VOC levels rather than ETS.

Although RSP levels appeared to be influenced by smoking to a greater degree than VOC levels, the contribution of ETS to total RSP was found to be low. Based upon solanesol measurements, the contribution of smoking to indoor RSP levels ranged, on average, from 10 to 20%. RSP, solanesol and nicotine were the only constituents measurably influenced by smoking indoors, although the sum of other RSP sources clearly predominates. In Korean cities, the main sources of indoor RSP appear to be motor vehicles and industrial emissions, as well as heating and cooking, exacerbated by meteorological and locational factors.

The use of charcoal grills in Korean barbecue restaurants and unvented kerosene heaters in homes and offices was also found to have a significant adverse effect on the indoor levels of a number of pollutants, including RSP, CO and VOCs. The use of kerosene

heaters was one of the factors contributing to a marked seasonal increase in indoor pollution during the winter months. The other primary factor was vehicle emissions. It appears that poor operation of catalysts in cold weather combined with stagnant air conditions contributed to elevate VOC levels both indoors and outdoors.

Acknowledgements—The authors would like to thank Professor Yashushi Kodama of The University of Occupational and Environmental Health in Japan for his valuable comments throughout this study. They would also like to thank Mr Y.M. Kim of Yeungnam University and Mr Y.H. Yoon of Hanyang University for their assistance in connection with the sampling and analytical work involved in this study, and Mr Ivan Gee, Centre for Environmental Control and Waste Management, Imperial College, for his help with the manuscript. The authors are also grateful to the Korea Research Institute of Chemical Technology for their participation in nicotine interlaboratory comparisons. Finally, the authors would like to thank the Japan Indoor Air Research Society for providing the funding needed for this study.

REFERENCES

- ACS Committee on Environmental Improvement (1980) Guidelines for data acquisition and data quality evaluation in environmental chemistry. *Anal. Chem.* **52**, 2242–2249.
- Baek S. O. (1993) Legislative aspects of the emission control of hydrocarbons and other air pollutants in Korea. In *Volatile Organic Compounds in the Environments* (edited by Leslie G. and Perry R.), pp. 33–42, Lonsdale Press, Indoor Air International, London.
- Bevan M. A. J., Proctor C. J., Baker-Rogers J. and Warren D. (1991) Exposure to carbon monoxide, respirable suspended particulates, and volatile organic compounds while commuting by bicycle. *Environ. Sci. Technol.* **25**, 788–791.
- Black F. M., High L. E. and Lang J. M. (1980) Composition of automobile evaporative and tailpipe hydrocarbon emissions. *J. Air Pollut. Control Ass.* **30**, 1216–1221.
- Cha C. W. and Cho S. H. (1990) Characterization of indoor air pollution in Korea. In *Indoor Air Quality* (edited by Kasuga H.), pp. 442–447, Springer, Berlin.
- Chan C. C., Ozkaynak H., Spengler J. D. and Sheldon L. (1991) Driver exposure to volatile organic compounds, CO, ozone and NO₂ under different driving conditions. *Environ. Sci. Technol.* **25**, 964–972.
- Cohen M. A., Ryan P. B., Yanagisawa Y., Spengler J. D., Ozkaynak H. and Epstein P. S. (1989) Indoor/outdoor measurements of volatile organic compounds in the Kanawha Valley of West Virginia. *J. Air Pollut. Control Ass.* **39**, 1086–1093.
- Connor J. M., Oldaker G. B. III. and Murphy J. J. (1990) Method for assessing the contribution of environmental tobacco smoke to respirable suspended particles in indoor environments. *Environ. Technol.* **11**, 189–196.
- Crump D. R. and Madany I. M. (1993) Daily variations of volatile organic compound concentrations in residential indoor air. *Proc. Indoor Air '93*, **2**, 15–20.
- Daisey J. M., Hodgson A. T., Fisk W. J., Mendell M. J. and Ten Brinke J. (1994) Volatile organic compounds in twelve California office buildings: classes, concentrations and sources. *Atmospheric Environment* **28**, 3557–3562.
- Dann T. F. and Wang D. K. (1995) Ambient air benzene concentrations in Canada (1989–1993): seasonal and day of week variations, trends, and source influences. *J. Air Waste Man. Ass.* **45**, 695–702.

- Edgerton S. A., Holdren M. W., Smith D. L. and Shah J. J. (1989) Inter urban comparison of ambient volatile organic compound concentrations in U.S. cities. *J. Air Waste Man. Ass.* **39**, 729-732.
- Evans G. F., Lumpkin T. A., Smith D. L. and Somerville M. C. (1992) Measurements of VOCs from the TAMS network. *J. Air Waste Man. Ass.* **42**, 1319-1323.
- Fellin P. and Otson R. (1994) Assessment of the influence of climatic factors on concentration levels of volatile organic compounds (VOCs) in Canadian homes. *Atmospheric Environment* **28**, 3581-3586.
- Feron V. J., Til H. P., Vrijer F. and van Bladeren P. J. (1992) Toxicology of volatile organic compounds in indoor air and strategy for future research. *Indoor Envir.* **1**, 69-81.
- Field R. A., Goldstone M. E., Lester J. N. and Perry R. (1992) The sources and behaviour of tropospheric anthropogenic volatile organic compounds. *Atmospheric Environment* **26A**, 2983-2996.
- Hartwell T. D., Pellizzari E. D., Perritt R. L., Whitmore R. W. and Zelon H. S. (1987) Comparison of volatile organic levels between sites and seasons for the total exposure assessment methodology (TEAM) study. *Atmospheric Environment* **21**, 2413-2424.
- Heavner D. L., Morgan W. T. and Ogden M. W. (1994) Determination of volatile organic compounds and ETS apportionment in 49 homes. *Envir. Int.* **21**, 3-21.
- Holcomb L. C. and Seabrook B. S. (1995) Indoor concentrations of volatile organic compounds: implications for comfort, health and regulation. *Indoor Envir.* **4**, 7-26.
- Hornung R. W. and Reed L. D. (1990) Estimation of average concentration in the presence of non-detectable values. *Appl. Occup. Envir. Hyg.* **5**, 46-51.
- Kawamoto T., Matsuno K., Arashidani K., Yoshikawa M., Kayama F. and Kodama Y. (1993) Personal exposure to nitrogen dioxide from indoor heaters and cooking stoves. *Arch. Envir. Contam. Toxicol.* **25**, 534-538.
- Kim Y. S. (1992) A literature review on indoor air quality in Korea. *J. Kor. Pub. Health Ass.* **18**, 147-165.
- Kirk P. W. W., Hunter M., Back S. O., Lester J. N. and Perry R. (1988) Environmental tobacco smoke in indoor air. In *Indoor and Ambient Air Quality* (edited by Perry R. and Kirk P. W. W.), pp. 99-112. Selver Ltd, London.
- Kodama Y., Arashidani K., Kawamoto T., Matsuno K., Yoshikawa M., Kayama F. and Hirai M. (1991) Indoor pollution by oil heaters and cigarette smoke. *Toxicol. Ind. Hlth.* **7**, 169-177.
- Korea Environmental Science Research Council (1989) Research on Indoor Air Pollution and Toxic Air Pollutants. 150 pp. (in Korean).
- Kostianen R. (1995) Volatile organic compounds in the indoor air of normal and sick house. *Atmospheric Environment* **29**, 693-702.
- Lee K. H., Lee Y. G., Moon E. M. and Lim J. G. (1995) Indoor air quality and sick building syndrome in winter office building condition in Korea. In *Proc. Int. Conf. on Indoor Air Quality in Practice*, Oslo, Norwegian Society of Chartered Engineers and International Society of Indoor Air Quality and Climate, pp. 288-299.
- Leslie G. B. and Haraprasad V. (1993) Indoor air pollution from combustion sources in developing countries. *Indoor Envir.* **2**, 4-13.
- Ogden M. W., Eudy J. W., Heavner D. L., Conrad F. W. and Green C. R. (1989) Improved gas chromatographic determination of nicotine in environmental tobacco smoke. *Analyst* **114**, 1005-1008.
- Ogden M. W., Maiolo K. C., Oldaker G. B. and Conrad F. W. (1990) Evaluation of methods for estimating the contribution of ETS respirable suspended particles. In *Proc. 5th Int. Conf. on Indoor Air Quality and Climate*, Toronto, Indoor Air 90, Vol. 2, pp. 415-420.
- Ostoson R., Fellin P. and Tran Q. (1994) VOCs in representative Canadian residences. *Atmospheric Environment* **28**, 3563-3569.
- Perry R. and Gee I. L. (1994) Vehicle emissions and effects on air quality: indoors and outdoors. *Indoor Envir.* **3**, 224-236.
- Phillips K., Howard D. A., Browne D. and Lewsley J. M. (1994) Assessment of personal exposures to environmental tobacco smoke in British nonsmokers. *Envir. Int.* **20**, 693-712.
- Rasmussen R. A. and Khalil M. A. K. (1983) Atmospheric benzene and toluene. *Geophys. Res. Lett.* **10**, 1096-1099.
- Scheff P. A., Wadden R. A., Bates B. A. and Aronson P. F. (1989) Source fingerprints for receptor modeling of volatile organics. *J. Air Pollut. Control Ass.* **39**, 469-478.
- Shah J. J. and Singh H. B. (1988) Distribution of volatile organic chemicals in outdoor and indoor air. *Envir. Sci. Technol.* **22**, 1381-1388.
- Singh H. B., Salas L., Vezee W., Sittou B. and Ferek R. (1992) Measurement of volatile organic chemicals at selected sites in California. *Atmospheric Environment* **26A**, 2929-2946.
- Son B. S., Nitta H., Maeda K., Kim Y. S. and Yanagisawa Y. (1990) Measurements of indoor CO concentrations and personal exposures in Korea. *J. Japan Soc. Air Pollut.* **25**, 334-342.
- Sweet C. W. and Vermette S. J. (1991) Toxic volatile organic compounds in urban air in Illinois. *Envir. Sci. Technol.* **26**, 165-173.
- Tancredi M., Wilson R., Zeise L. and Crouch E. A. C. (1987) The carcinogenic risk of some organic vapors indoors: a theoretical survey. *Atmospheric Environment* **21**, 2187-2205.
- Tille K. J. W., Savelsberg M. and Bachmann K. (1985) Airborne measurements of nonmethane hydrocarbons over Western Europe: vertical distributions, seasonal cycles of mixing ratios and source strengths. *Atmospheric Environment* **19**, 1751-1706.
- U.K. Department of Environment (1994) Expert Panel on Air Quality Standards: Benzene, London HMSO.
- Yanagisawa Y. and Nishimura H. (1980) A personal sampler for measurement of nitrogen dioxide in ambient air. *J. Japan Soc. Air Pollut.* **15**, 316-323.
- Yocom J. E. (1982) Indoor-outdoor air quality relationships — a critical review. *J. Air Pollut. Control Ass.* **32**, 500-520.